

Review

Research Progress on Chiral Supramolecular Sensors for Enantiomer Detection

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Abstract: Chiral substances occur naturally in abiotic and living systems. The recognition and detection of chiral substances in the natural environment or their analysis and detection in biological systems are crucial. Chiral recognition is a research hotspot in clinical medicine, pharmacology, biochemistry, and other fields. Indeed, many researchers have developed various sensors with different functionalized materials for detecting and analyzing enantiomers. Supramolecular systems have important applications in the development of molecular recognition technologies, and the development of supramolecular chemistry is closely related to research on molecular devices. Therefore, this review summarizes the principle of chiral supramolecular sensors for the detection of enantiomers from the perspective of various sensor types, including optical, electrochemical, electrochemical luminescence, photoelectric, and supramolecular chemical sensors. This review also summarizes the relevant reports on chiral supramolecular sensors in the last five years. Finally, we highlight the prospects of supramolecular chiral sensors in future research.

Keywords: chiral matter; optics; electrochemistry; electrochemical luminescence; photoelectric chemistry; chiral supramolecular sensors



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1. Introduction

Chiral compounds occur naturally in living organisms. In general, naturally occurring enantiomers have no adverse effects on the environment and living organisms. However, several enantiomers present in the ecosystem can pollute the environment and cause health risks [1–3]. Chiral substances, similar to invasion by foreign bodies, have become a threat in terms of tracing their degradation and particularly, for example, in the contamination of chiral drugs [4,5], as most drugs have chiral configurations. Although the other isomeric configuration in chiral drugs has opposite toxicological and pharmacokinetic properties, it has the same physicochemical properties [6]. As a result, chiral drugs containing hazardous substances can enter the natural environment during production, leading to considerable damage to the environmental matrix and uncontrollable risks [7]. Therefore, monitoring and analyzing chiral substances in recycled wastewater and biosolids is indispensable [8,9].

Chiral drugs have potential application values in clinical medicine. In particular, chiral compounds are the active ingredients in some traditional Chinese medicine formulations [10], but the purification process of these natural drugs is complex and tedious; therefore, only pure chiral compounds with medicinal values are synthesized. Among the different chiral compounds, small chiral molecules such as amino acids are intermediates or chiral inducers of many chiral drugs [11,12]. Thus, the specific recognition of certain amino acid enantiomers is necessary. In addition, due to technological limitations and cost, several chiral drugs are synthesized in the form of a racemic mixture [13]. However, since the pharmacological activities of the two isomers of chiral drugs are different, it is necessary to mitigate undesirable physiological effects through the cyclic metabolism of

the toxic structure in the enantiomer. Regardless of the chiral drugs used to treat diseases, it is crucial to detect chiral drug residues in organisms [14]. Furthermore, the hormones secreted by living organisms are chiral structures, and the specific recognition of these chiral molecules is required to interpret routine results of vital signs and implement dynamic monitoring [15,16].

Supramolecular chemistry involves multiple scientific fields and is an interdisciplinary science involving chemistry, biology, physics, materials science, and other disciplines. Since supramolecular chemistry was proposed about 55 years ago, research in this scientific field has mainly focused on host–guest chemistry and ordered supramolecular assemblies [17,18]. In the 1990s, Silva et al. combined the concepts of host–guest chemistry and fluorescence and applied supramolecular chemistry in the field of optical sensing to detect ions in blood [19]. However, in recent years, researchers have focused on the molecular recognition of supramolecular species and directly used the interaction of supermolecules to build sensors [20]. Molecular recognition is the selective binding of the subject (receptor) to the object (ligand) to produce certain specific functions [21]. The host–guest interactions mainly include hydrophobic interaction, hydrogen bonding, electrostatic interaction, π – π stacking, and ion–dipole interaction. Among the various supramolecular compounds, macrocyclic compounds are often used as classical host molecules to recognize ligand molecules due to their cage cavity advantageous [22]. Macrocyclic compounds mainly include cyclodextrin, cucurbituril, pillararene, calixarenes, and crown ether. Moreover, due to their size and complexity, macrocyclic compounds exhibit larger spatial distributions to achieve host–guest interaction. Thus, the binding affinity and selectivity can be increased [22]. Through further examinations, a series of excellent host molecules were developed based on supramolecular interaction and used as sensors to achieve fast and simple chiral molecular recognition [23,24]. They include supramolecular self-assembly [25–27], supramolecular polymer [28,29], metal–organic complexes [30], supramolecular gels [31], chiral porphyrinoids [32], conducting polymer [33], molecularly imprinted polymer [34], and acyclic cucurbit[n]urils and molecular tweezers [35].

Chiral compounds have been detected and identified using traditional methods, including liquid chromatography [36] and mass spectrometry [37], which require complex sample pretreatment and tedious operation of instruments. Supramolecular chiral sensors can be used to rapidly and sensitively analyze and detect chiral compounds without complex preliminary work through the combined use of instruments and can achieve real-time data monitoring [38]. Supramolecular chiral sensors are classified into optical, electrochemical, electrochemical luminescence, and photoelectrochemical (PEC) sensors. This article reviews the relevant literature on chiral supramolecular sensors used in enantiomer detection and analysis over the past five years. Supramolecular systems play a crucial role in the development of molecular recognition technologies. Among the different analytical methods, the optical analysis method has the longest history of detailed studies and numerous advantages. The electrochemical analysis method has played an important role in chiral molecular recognition. Although there are few methods using electrochemiluminescence (ECL) and PEC analysis, most of them have achieved better results. All types of chiral supramolecular sensors were constructed based on the functional design of receptor molecules and host–guest interactions to achieve the molecular recognition of enantiomers. Therefore, it is essential to study auxiliary materials to improve the sensitivity, stability, and reproducibility of the sensor.

2. Optical Chiral Supramolecular Sensor

Spectroscopy has broad application prospects in chiral research and chiral drug analysis [39,40]. Optical fluorescence sensors have the advantages of high sensitivity, good selectivity, less sampling requirement, and are not affected by external electromagnetic fields [41]. Moreover, large cyclic compounds with conjugate structures are suitable for modifying fluorescent probes [42], and most biomolecules exhibit photoluminescent prop-

erties [43]. Therefore, supramolecular fluorescent transducers have wide applications in chiral substance recognition [44].

The absolute adherence of circular dichroism to the structure of chiral substances enables the high-throughput screening of chiral substances. Additionally, chiral macrocyclic compounds or self-assembled chiral hosts are used for the specific recognition of chiral molecules through host–guest interactions [45]. In recent years, the emergence of a new concept, namely “chiral induced amplification”, has made the construction of chiral sensors simpler and more diversified [46–52], thus enabling chiral amplification [53–55] or chiral transfer [56,57] to the host of the non-chiral cage through chiral guest molecules.

In addition to the above-mentioned optical sensors, other methods have been used, such as a colorimetric method based on the Lambert–Beer law, a room-temperature phosphorescence method, and a new surface plasmon resonance (SPR) method, in part of the construction and application of supramolecular chiral sensors. Colorimetric sensors are characterized by desirable selectivity, high stability, and low cost, and can be used as paper-based sensors [58]. The room-temperature phosphorescence method is characterized by high sensitivity, wide linear range, simple operation, and fast detection [59]. SPR sensors have high sensitivity and can achieve real-time and unlabeled detection [60].

2.1. Fluorescence Sensor for the Detection of Enantiomers

Recently, compared with traditional fluorescent nanoprobe, the new fluorescent nanoprobe based on noble metal nanoclusters and nanocomposites are characterized by excellent luminescence performance, simple and controllable preparation, easy functionalization, and good biocompatibility. In 2018, Chen et al. constructed papain-stabilized gold nanoclusters (papain@AuNCs) and then complexed the fluorescently quenched Cu^{2+} to form fluorescent probes (Figure 1) [61]. Since the nanomaterials generated using D-penicillamine easily form complexes with Cu^{2+} , the fluorescence signal response is recovered, and a highly sensitive chiral sensor to recognize D-penicillamine can be constructed. Protease substances are added to the sensor to enhance the biocompatibility of the probe. In 2018, Yu et al. prepared 11-mercaptopropionic acid (MUA)-capped gold nanoclusters (AuNCs@MUA) with good water solubility [62] and also constructed a Cu^{2+} -switched fluorescent probe to detect penicillamine, which exhibits higher photostability and quantum yield than other Cu^{2+} coordination compounds. In 2021, Liu et al. constructed a fluorescence sensor to detect D-penicillamine and also prepared a composite material composed of DNA-templated silver nanoclusters (DNA-AgNCs) [63], which can identify D-penicillamine via aggregation–fluorescence quenching. The detection method using precious metal as a medium has been shown to be faster and more environmentally friendly. In 2018, Yuan et al. labeled a DNA sequence with carboxyfluorescein (FAM), and then functionalized gold nanoparticles (AuNPs) to form composite nanomaterials (aptamer-AuNPs), and quenched the fluorescence of aptamers [64]. The subsequent addition of arginine enantiomers to the specific binding of aptamer resulted in varying degrees of aptamers released from AuNPs, and corresponding fluorescence intensity signals were detected. The chiral fluorescence sensor constructed using this method could specifically detect and identify both L-arginine and D-arginine. This sensor uses DNA sequences as functional materials to reduce chemical contamination. In 2021, Huy et al. prepared composite nanomaterials by functionalizing CdTe quantum dots with β -cyclodextrin [65], where the cavity of β -cyclodextrin provides an excellent environment for supramolecular interaction with aspartic acid, thereby promoting the fluorescence quenching of the receptor to achieve the selective quantitative detection of aspartic acid. Therefore, this composite material can be used to obtain a sensor that is simple and easy to build.

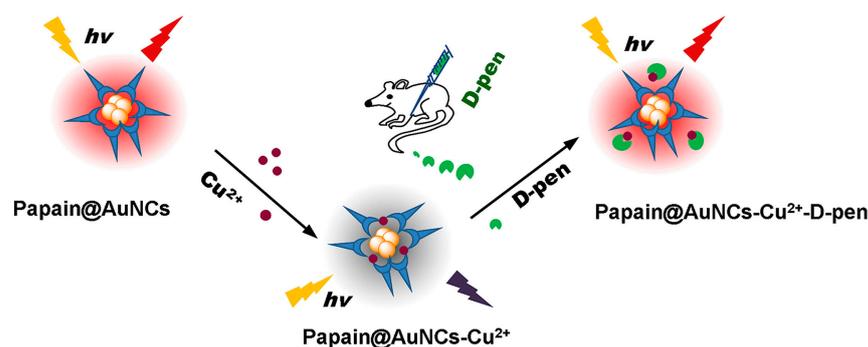


Figure 1. Schematic diagram of Papain@AuNCs- Cu^{2+} sensor recognition of D-penicillamine. Adapted from Ref. [61]. Copyright 2018 ELSEVIER.

Compared with traditional labeling materials, quantum dots are characterized by high fluorescence intensity, diverse luminous colors, and a wide spectrum range. Moreover, quantum dots are suitable for multiple spatial and spectral transmission, effective attenuation, and elimination of background fluorescence effects. Due to their superior properties, quantum dots have become highly effective fluorescent probes used for numerous applications. In 2019, Lu et al. constructed a fluorescence sensor to identify nitrophenol isomers using a chiral carbon-point hybrid porous organic cage nanocomposite (CD@RCC3) (Figure 2) [66]. This nanocomposite has a large void volume, stable fluorescence, and good solubility in organic reagents. In addition, the chiral isomers can be directly identified by simple host–guest interactions, and as a result, a quick and simple detection is achieved. In 2019, Masteri-Farahani et al. modified chiral colloidal CdSe quantum dots (CdSe-QDs) with cysteine enantiomers to regulate the optical properties of the quantum dot core and functionalize it [67]. The composite material has strong interactions with L-morphine and can quench its fluorescence, enabling the novel fluorescence sensor to selectively identify L-morphine.

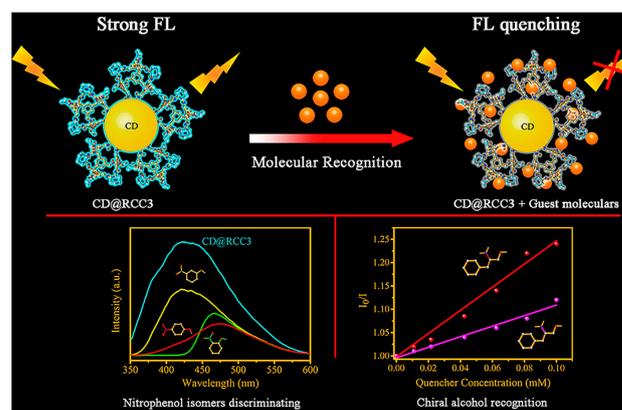


Figure 2. A fluorescence sensor constructed with CD@RCC3 was used to identify nitrophenol isomers. Adapted from Ref. [66]. Copyright 2019 ELSEVIER.

Metal–organic frameworks (MOFs) are novel supramolecular materials with large surface areas, abundant structures, and adjustable pores, and their abundant luminescence center has a good application prospect in fluorescence sensing analysis. In 2020, Xiao et al. functionalized the chiral S-1/R-1 UiO-66-(COOH)₂ with L/D-amino propanol to construct homochiral metal–organic frameworks (HMOFs) (Figure 3) [68]. The incompatibility of the HMOF assembly was simplified, and the fluorescence sensors constructed with S-1 (L-AP@UiO-66-(COOH)₂) and R-1 (D-AP@UiO-66-(COOH)₂), respectively, showed enhanced fluorescence signal intensity with the phenylalanine enantiomer. This fluorescence sensor exhibits significant enantioselectivity.

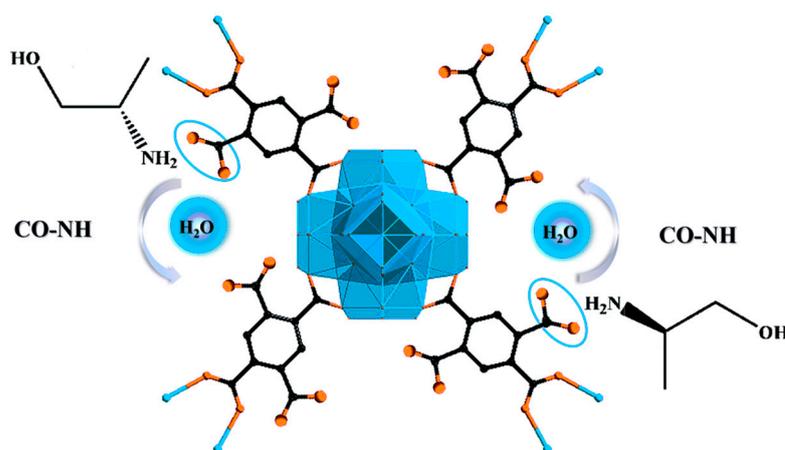


Figure 3. Composite material design diagram.

Coordination polymers have spatial advantages similar to those of MOFs and are excellent hosts for supramolecular compounds. In 2022, Thoonen et al. designed and synthesized a novel topological coordination polymer using 1,1'-bi-2-naphthol (BINOL), trimesic acid, and Zn^{2+} [69]. The polymer exhibited an excellent chiral three-dimensional (3D) spatial structure. Through fluorescence quenching experiments on selected chiral analytes, the constructed fluorescence sensor showed high enantioselectivity for a series of chiral compounds. In 2022, Tang et al. used a simple hydrothermal method to synthesize the CdTe quantum dots of fluorescent molecularly imprinted polymer (MIP) particles (CdTe@MIPs) and self-absorbed them into the activated capillary tube (Figure 4) [70]. The interaction between the infrared capillary sensor and L-histidine enhanced the fluorescence of the material. Experiments showed that this fluorescence sensor can be used to determine L-histidine concentration in real samples. Therefore, the trace analysis of target molecules can be achieved.

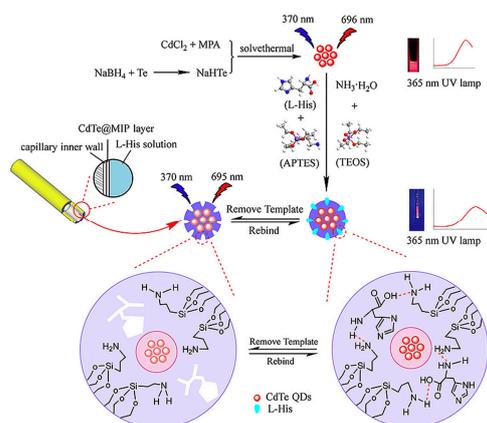


Figure 4. CdTe@MIPs were synthesized to construct chiral sensors for L-histidine recognition. Adapted from Ref. [70]. Copyright 2022 ELSEVIER.

It is well known that nucleic acids with high affinity and specificity can be used as chiral aptamers to bind to target substrates (through base pairing), so as to achieve chiral recognition. In 2019, Chovelon et al. reported the measurement of enantiomers using the aptamer kissing complex (AKC) (Figure 5) [71]. In this case, the arginine vasopressin enantiomers processed L/D-DNA as an aptaswitch and L/D-RNA labeled with Texas red and luciferin as hairpin probes (aptakiss). As shown by the results, the fluorescence anisotropy signal was enhanced when the aptaswitch and the hairpin loop-loop interacted. The sensor simultaneously monitored L/D-arginine vasopressin and detected enantiomer impurities as low as 0.01%. This study successfully extended the range of kiss assembly

strategy to DNA-based chiral analysis and provided a very valuable strategy for the construction of supramolecular chiral sensors.

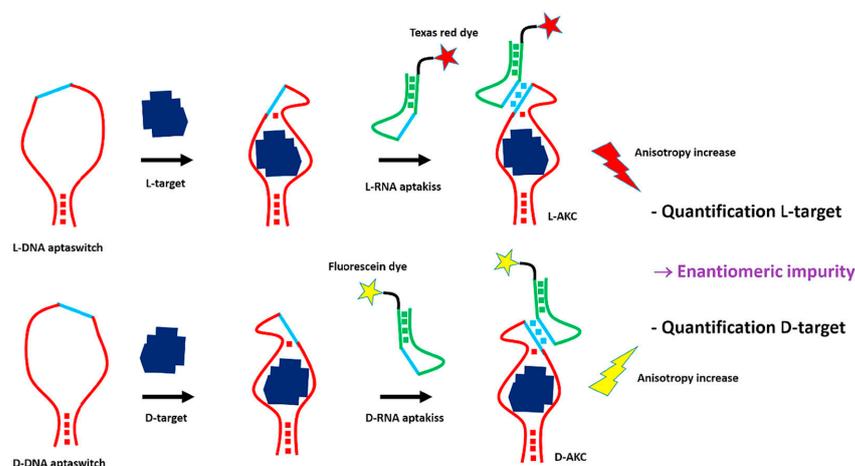


Figure 5. Schematic diagram of AKC FA chiral sensor. Adapted from Ref. [71]. Copyright 2019 ELSEVIER.

2.2. Circular Dichroic Sensor for the Detection of Enantiomers

The synthesis of chiral macrocyclic compounds has been a challenge because the various synthesis methods are still limited by low yield, unsatisfactory stereoselectivity, and the use of equivalent chiral accelerators. In 2019, Ohishi et al. developed a BINOL-derived chiral macrocycle for the chiral recognition of native saccharides [72]. The experimental process of specific recognition was completed through the measurement of the circular dichroism spectrum; therefore, chiral macrocyclic compounds are used in circular dichroism sensors for more highly selective detection. In 2020, Chai et al. synthesized a new water-soluble chiral macrocyclic compound, a chiral naphthalene tube [73]. The hydrophobic cavity of this macrocyclic compound has a different affinity for chiral neutral molecules; thus, it has a certain application prospect in terms of biological compatibility. Supramolecular compounds are used in the construction of circular dichroism sensors to achieve ideal selective enantiomeric recognition. In 2020, Huang et al. inserted secondary amine groups into non-chiral naphthalene tubes with hydrophobic cavities (Figure 6). The functionalized large rings were shown to be able to selectively identify chiral substances such as carboxylic acids in an aqueous environment through the non-covalent bonding of the salt bridge effect and showed different signal responses in the form of chiral transfer for circular dichroism detection [74].

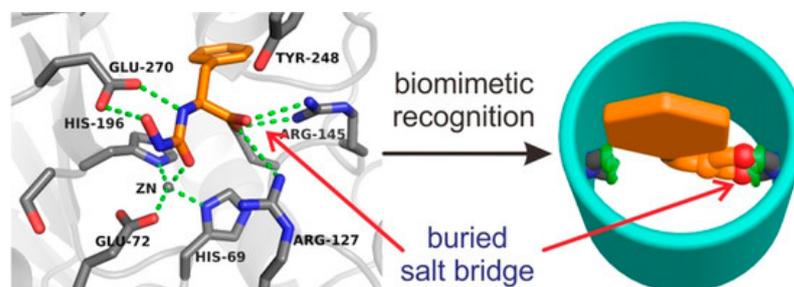


Figure 6. Diagram of embedding a secondary amine group in a non-chiral naphthalene tube. Adapted from Ref. [74]. Copyright 2020 Wiley Online Library.

Currently, chiral nanomaterials are mainly fabricated via the introduction of chiral ligands, the construction of helical structures, and other electric dipole moment control methods. However, due to their environmental stability and electrical conductivity, chiral materials have limited practical applications. Therefore, exploring new regulatory mechanisms and constructing novel chiral nanofunctional materials is challenging. In 2018,

Liu et al. used poly(2-oxazoline) to self-assemble chiral nanomicelles (CPOx2) with a strong circular dichroism signal [75]. The amphiphilic copolymer was used as a chiral probe to selectively detect histidine enantiomers in aqueous solutions. In 2019, Guo et al. synthesized a pair of water-soluble chiral 2,6-helic [6] arene derivatives, which was used as a supramolecular assembly with 4-[(4'-n, N-diphenylamino)styryl]-n-methylpyridinium iodide in a one-to-one host-guest interaction, showing that the chirality of the receptor is continuously transferred to an achiral guest molecule [76]. This supramolecular material has certain application potentials in the construction of chiral circular dichroism sensors. In 2019, Bao et al. independently constructed a chiral supramolecular pump containing aromatic compounds (Figure 7) [77]. The aromatic compounds were 3D-rotated into left-handed and right-handed solid fibers with significant circular dichroism signals. The abundant tubular pores of the supramolecular pump can absorb a large number of enantiomer compounds to purify chiral pollutants in an aqueous environment. In 2023, Hirao et al. developed supramolecular helical polymers constructed using a tetrakis (porphyrin) frame with different branched chains, which produced obvious electronic circular dichroism (ECD) signals, to build supramolecular chiral sensors for pinene enantiomers [78]. This finding contributed to the establishment of supramolecular chiral sensors for pure hydrocarbons based on ECD probes, and few cases have been presented to date.

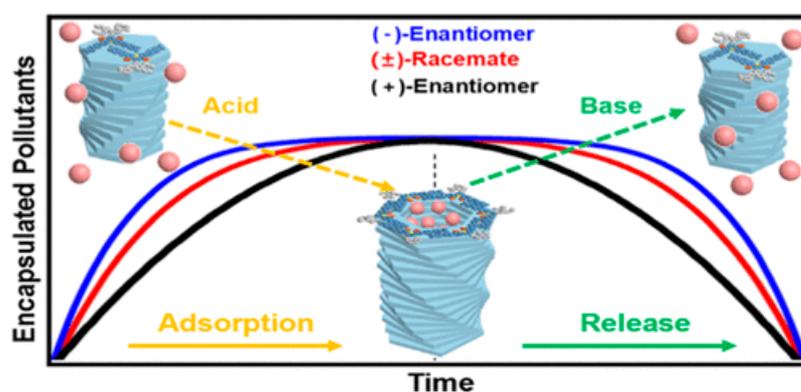


Figure 7. Schematic diagram of a chiral supramolecular pump with an aromatic compound. Adapted from Ref. [77]. Copyright 2019 ACS.

2.3. Other Optical Sensors for the Detection of Enantiomers

Besides the two reported optical sensors, we reviewed research on intuitive colorimetric and room-temperature phosphorescent sensors. In 2018, Zor et al. prepared inherently chiral silver nanopolymers by embedding silver nanoparticles (AgNPs) into nanopaper, which were developed into disposable cubets for the selective recognition and detection of D-cysteine [79]. In 2019, Copur et al. used L-cysteine-functionalized carbon quantum dots to synthesize chiral carbon quantum dots, which showed strong phosphorescence and could interact with L-lysine [80]. Eventually, the material was developed into a disposable nanopaper-based chiral sensor to recognize lysine enantiomers (Figure 8). The disposable chiral sensor is characterized as a low-cost, simple process, with high sensitivity and high practical application value.

In addition, SPR is an optical sensing technique commonly used for biological analysis. In most cases, SPR does not require sample pretreatment and dynamically monitors biomolecular interactions without the need for labeled samples. In 2018, Xu et al. used L-tryptophan as a template to crosslink graphene oxide (GO) with self-polymerized dopamine to prepare a MIP and modified the composite material (Gr/MIP) on the modified chip to produce an SPR sensor to recognize tryptophan enantiomers [81].

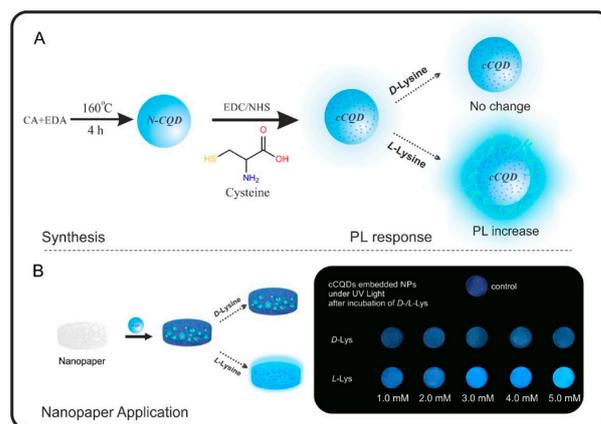


Figure 8. Schematic diagram of the designed chiral nanopaper sensor for selective recognition of lysine enantiomers. Adapted from Ref. [80]. Copyright 2019 ELSEVIER.

3. Electrochemical Chiral Supramolecular Sensors

An electroanalytical method is a fast, sensitive, and accurate method for trace analysis. Electrochemical sensors in analytical chemistry are usually based on a three-electrode system used to detect target molecules. These electrode processes include the chemical steps that occur, new phase generation, and surface diffusion steps in a solution or on the electrode surface. The unique molecular structure of supramolecules can be combined with numerous organic, inorganic, and biological molecules to form inclusion complexes on the electrode surface or in the solution for molecular recognition and the selective pre-enrichment of analytes [82], which can be used to build new molecular selective electrochemical sensors. A series of chiral electrochemical sensors with supramolecular compounds as the core device are constructed using the interaction and inclusion mode between the achiral host compounds and chiral guest molecule [83,84], thus improving the sensitivity and selectivity of the sensors and providing multivariate and rapid detection methods for identifying chiral compounds [85–88]. Among the various sensors, optimized electrode modification materials and a functionalized supramolecular host can be used in modified chiral electrochemical sensors. The methods of electrochemical determination based on relevant reports are cyclic voltammetry and differential pulse voltammetry with high enantioselectivity.

3.1. Enantioselectivity Using Cyclic Voltammetry

Most nanomaterials can increase electron transfer rates to enhance the conductivity of the environment; thus, various functional nanocomposites have been synthesized to optimize the electrochemical sensor. In 2018, Munoz et al. constructed a chiral electrochemical sensor by solidifying a modified chiral magnetic nanobiofluid onto a nanocomposite graphene paste electrode (Figure 9) [89]. They obtained magnetic nanobiofluids (mNBFs) by loading β -cyclodextrins on AuNPs modified with cobalt ferrite nanoparticles. The interaction between tryptophan enantiomers and mNBFs can generate different cyclic voltammetry oxidation potentials and thus L/D-Trp can be identified. In 2020, Zhou et al. modified AgNPs onto gold microelectrodes for host–guest interaction with cysteine enantiomers, achieving different degrees of dynamic coupling for selective detections [90]. The gold-coated glass nanofibers form a new sensing interface, which serves as a new guideline for constructing chiral electrochemical sensors. In 2021, Yang et al. constructed a disposable chiral electrochemical sensor using L-cysteine-functionalized carbon nanotube-modified screen-printed electrodes [91]. The cyclic voltammetry signal of S-mandelic acid was significantly higher than that of R-mandelic acid, thus achieving the selective recognition of mandelic acid enantiomers. The sensor based on screen printing electrode is characterized by low cost and potential commercial applications. In 2021, He et al. used aniline to synthesize S/R-polyaniline with a bond shape using camphor sulfonic acid enantiomers,

which are used for the highly selective recognition of tryptophan enantiomers solidified on the electrode [92]. This chiral polymer material is easily synthesized and widely used.

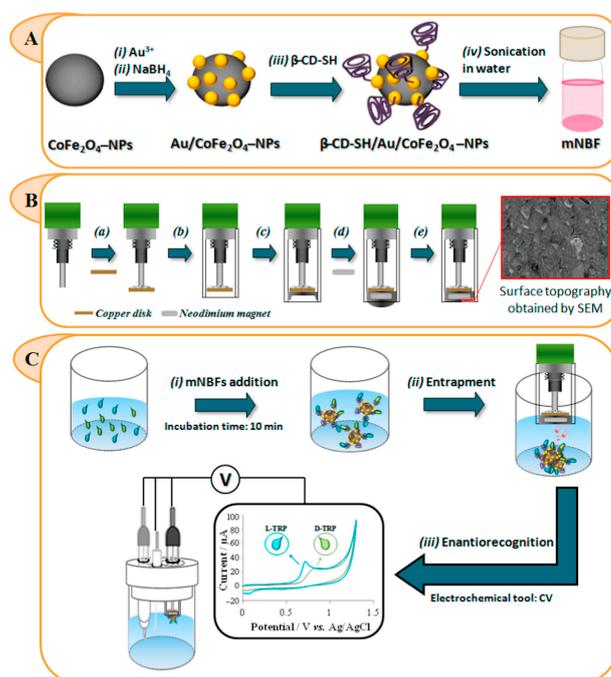


Figure 9. Schematic diagram of chiral electrochemical sensor construction ((A–C) represents the response signal of the sensor to the cyclic voltammetry test of tryptophan enantiomer.). Adapted from Ref. [89]. Copyright 2018 ELSEVIER.

3.2. Enantioselectivity Using Differential Pulse Voltammetry

Like the specific recognition mechanism of biological antibodies and antigens, molecular imprinting technology is used to prepare synthetic receptors for detecting target molecules, namely, MIPs [93]. Compared with natural biomolecular recognition systems such as monoclonal antibodies or receptors, MIPs are characterized by unique physical, chemical, and mechanical properties, specific selectivity, high stability, and simple preparation [94]. Therefore, molecular imprinting is widely used in molecular recognition using electrochemical sensors. In 2019, Duan et al. used a Prussian blue-functionalized carbon-nanotube-modified electrode and then electropolymerized pyrrole on this composite material using cysteine as a template to prepare MIPs [95]. The constructed electrochemical sensor can selectively recognize cysteine enantiomers and detect dopamine in human serum and pollutants in the environment. In 2020, Zhang et al. modified double-stranded DNA as a functional unit on the gold electrode and then inserted D-carnitine to prepare a MIP template (Figure 10) and showed that the cavity of MIPs had higher specificity [96]. The constructed electrochemical sensor enabled the selective ultra-trace determination of D-carnitine in carnitine enantiomers. In 2022, Karadurmus et al. modified β -cyclodextrin on a glass carbon electrode as a functional unit; then, they incorporated esomeprazole through supramolecular interactions to obtain a MIP template and ultimately prepared a 3D porous network with ethyl orthosilicate on the electrode surface to provide more binding sites [97]. The sensor was used for the selective detection of esomeprazole enantiomers in a serum environment. In 2022, Karadurmus et al. also reported the selective recognition of pseudoephedrine enantiomers with MIPs prepared via the in situ electropolymerization of o-phenylenediamine [98], and this electrochemical sensor has a high recovery rate in pharmaceutical formulations and serum environments.

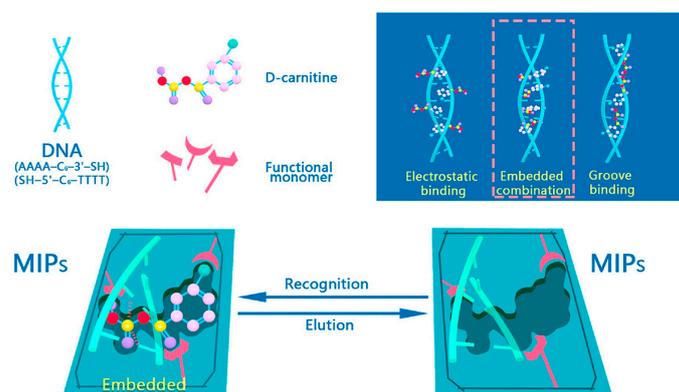


Figure 10. Schematic diagram of a template for a molecularly imprinted polymer. Adapted from Ref. [96]. Copyright 2020 ELSEVIER.

Macrocyclic compounds are excellent receptor molecules used to produce supramolecular effects. Among the various macrocyclic compounds, cyclodextrin has raised significant research interest among many researchers due to its excellent host molecule properties similar to those of enzymes. Various organic compounds can be embedded in the hydrophobic cavity of cyclodextrin to form an inclusion complex, which changes the physical and chemical properties of the envelope [99]. Therefore, it is possible to crosslink cyclodextrin molecules with various functional groups or polymers and perform chemical modification or polymerization using cyclodextrin as a monomer [100,101]. In 2018, Liang et al. constructed an electrochemical sensor on the electrode surface by coupling β -cyclodextrin to form a composite material on 3D graphene [102]. Then, the cavity of β -cyclodextrin was used as the binding site to establish host–guest interactions on the tryptophan enantiomer. As the binding force of L-tryptophan is much higher than that of D-tryptophan, the specific recognition of tryptophan enantiomers can be achieved. In 2019, Niu et al. modified β -cyclodextrin to form a composite material through the self-assembly of ferrocene and GO to construct an electrochemical sensor [103] and identified phenylalanine enantiomers through host–guest interactions, in which L-phenylalanine exhibited a higher affinity than D-phenylalanine. In addition, Cu^{2+} was independently assembled into a composite material after coordination with β -cyclodextrin and carboxymethyl cellulose, and it was fixed on the electrode modified with N-mixed 3D reduced GO to construct an electrochemical sensor [104] that recognized L/D-tryptophan through different degrees of binding. In 2019, Yi et al. also constructed an electrochemical sensor (Figure 11) by modifying carbon nanotubes coated with reduced GO on the electrode surface and then fixing β -cyclodextrin coated with rhodamine B on the electrode surface [105]. Through the competitive host–guest interaction between rhodamine B and phenylalanine enantiomers, β -cyclodextrin was easily bonded to L-phenylalanine; thus, the selective recognition of phenylalanine enantiomers was achieved. In 2020, Zou et al. prepared chiral composite materials with 6-O- α -maltose-functionalized β -cyclodextrin and fixed them on an electrode modified with a black phosphorus nanosheet [106]. The constructed sensor could recognize L/D-tyrosine enantiomers. In 2021, Gong et al. modified ammonium chitosan and carbon nanotubes on the electrode surface via self-assembly and then fixed Cu^{2+} on the electrode after coordination with β -cyclodextrin to create a sensing interface (Figure 12) [107]. They were able to selectively detect tryptophan enantiomers with the constructed electrochemical sensor. In 2021, Ebrahimi et al. reported the construction of an electrochemical sensor using β -cyclodextrin encapsulated in methylene blue composites [108], which could identify naproxen enantiomers through competitive host–guest interactions between methylene blue and the target molecule in chiral cavities. In 2022, Hou et al. reported a self-assembly formed by the electrooxidation of β -cyclodextrin on an electrode modified with chiral MOFs [109]. The constructed electrochemical sensor had multiple chiral recognition sites and could recognize tryptophan and penicillamine enantiomers. In 2022, Gao et al. synthesized a thiophene copolymer with cyclodextrin in a side chain (Figure 13) [110]. This

copolymer has good conductivity and chiral selectivity and can be used to construct an effective electrochemical chiral sensor to recognize amino acid enantiomers.

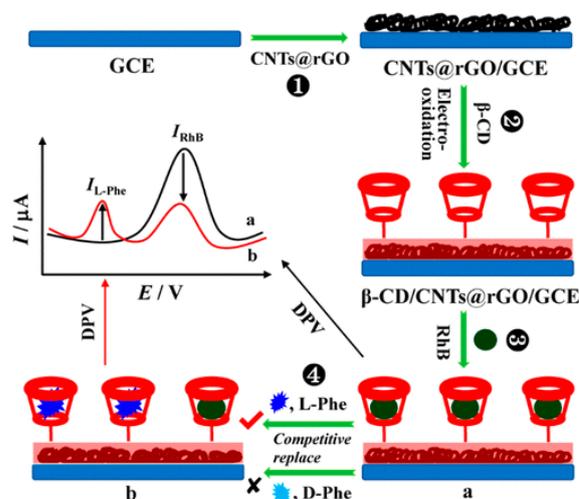


Figure 11. Diagram of recognition of phenylalanine enantiomers using a chiral sensor. Adapted from Ref. [105]. Copyright 2019 ACS.

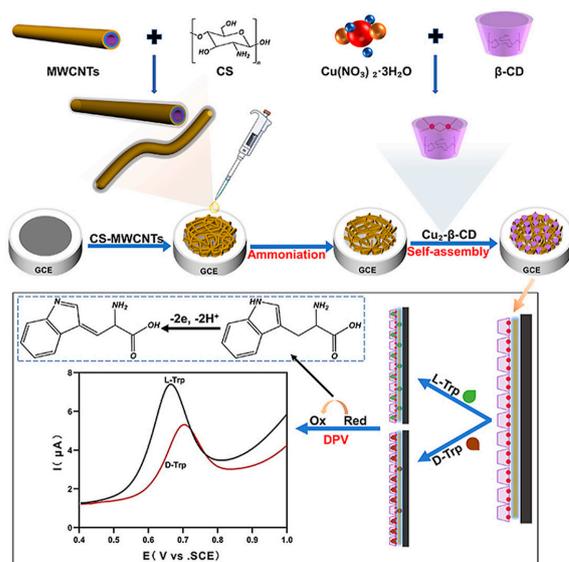


Figure 12. Diagram of chiral sensor recognizing tryptophan enantiomers. Adapted from Ref. [107]. Copyright 2021 ELSEVIER.

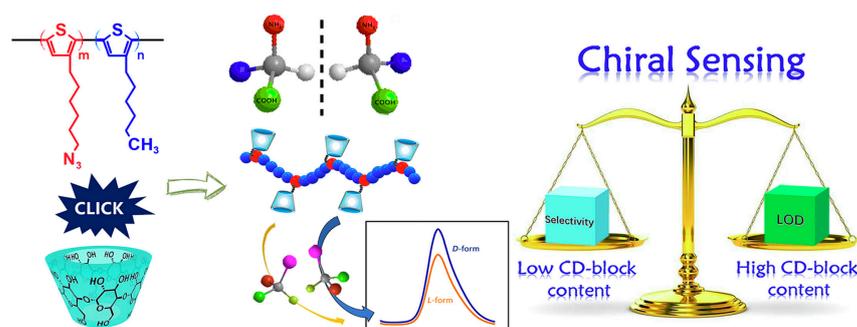


Figure 13. Schematic diagram of a chiral sensor constructed using thiophene copolymer with cyclodextrin in the side chain. Adapted from Ref. [110]. Copyright 2022 ELSEVIER.

Nanomaterials have been widely used in electrochemical sensing due to their unique properties, including excellent electrochemical performance, high surface areas, and adjustable conductivity. In 2018, Yang et al. used a 10-camphorsulfonic acid enantiomer for peroxidation polymerization to form polypyrrole, resulting in a spiral polypyrrole chain with a left-handed and right-handed chiral microenvironment [111]. The constructed electrochemical sensor can specifically recognize L/D-tryptophan enantiomers. In 2018, Zhang et al. used cellulose nanocrystals as chiral units functionalized with multiwalled carbon nanotubes to form a composite material to construct an electrochemical sensor [112]. The host-guest effect of the composite material on L-tryptophan was stronger than that of D-tryptophan, which allowed the sensor to selectively recognize and identify tryptophan enantiomers. In 2019, Pu et al. reported the enantiomeric separation of (6,5)-single-walled carbon nanotubes to construct a single chiral electrochemical sensor to selectively recognize 3, 4-dihydroxyphenylalanine enantiomers [113]. In 2019, Stoian et al. constructed an electrochemical sensor on a glassy carbon electrode modified with L-cysteine-functionalized AuNPs [114]. Their results showed that compared with a single AuNP compound, the functionalized nanocomposites enabled the selective recognition of the chiral drug propranolol as a result of the different degrees of host-guest interaction. In 2020, Lian et al. synthesized a dendritic gold nanomaterial [115] and used it to construct an electrochemical sensor that could amplify the chiral recognizability of guest molecules through a differential pulse method and specifically identify 3,4-dihydroxyphenylalanine enantiomers (Figure 14). In 2021, Jiang et al. introduced the polymerization of non-chiral phthalocyanine into L-lysine helical nanofibers to form a chiral microenvironment [116], and the electrochemical sensor constructed using the composite nanomaterials could identify tryptophan enantiomers. In 2021, Liu et al. self-assembled synthetic chiral ionic liquid and N-doped GO multiwall carbon nanotubes onto the electrode surface to prepare chiral composite nanomaterials [117]. Compared with the electrode without composite multiwall carbon nanotubes, the electrochemical sensor based on composite materials further amplified the chiral recognition signal of the target molecule for the specific detection of tryptophan enantiomers. In 2021, Yu et al. constructed a chiral composite material into N-acetyl-L-cysteine-functionalized pillar[5]arene, and then solidified the composite material onto a polycarbonate (PC) membrane to form nanochannels [118]. Ultimately, the modified PC membrane was fixed on a gold electrode to construct an electrochemical sensor (Figure 15). The chiral nanochannel had a stronger host-guest effect on S-propranolol that significantly reduced the electrochemical signal of the constructed sensor; however, it had no significant host-guest effect on R-propranolol and selectively recognized the propranolol enantiomer. In 2022, Li et al. synthesized helical AuNPs with intrinsic chirality to construct electrochemical chiral sensors [119]. L/D-helicoid Au nanoparticles specifically bound to L/D-tyrosine to produce an enhanced electrochemical signal.

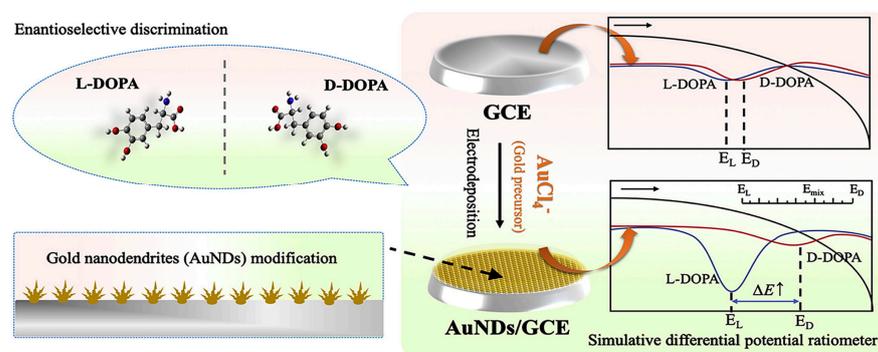


Figure 14. Schematic diagram of dendritic gold nanomaterials constructing an electrochemical sensor. Adapted from Ref. [115]. Copyright 2020 ELSEVIER.

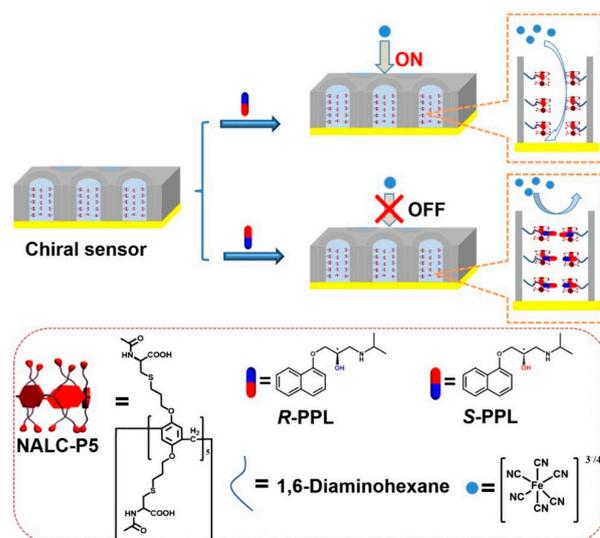


Figure 15. Schematic diagram of a composite solidified onto a PC film to form a nanochannel. Adapted from Ref. [118]. Copyright 2021 ELSEVIER.

4. Electrochemical Luminescence Chiral Supramolecular Sensors

ECL is a new analytical technique that combines electrochemistry and chemiluminescence for the detection of various analytes. ECL is characterized by high controllability, high sensitivity, and simplicity [120]. ECL mainly generates luminescence through an electrochemical reaction on the electrode surface to enable the quantitative and qualitative analysis of some specific compounds [121]. Although relevant research on ECL sensors has been conducted for nearly 50 years, due to the restriction of the electroluminescence of host molecules, it still faces certain challenges in the chiral molecular recognition field [122]. Few reports exist on chiral ECL supramolecular sensors, but reports on related studies are widely available.

Due to their physical and chemical properties, these nanomaterials have broad application prospects in chemical sensing, and their color and luminescence characteristics change with changes in particle sizes [123]. These characteristics and research findings can provide new guidelines to construct chiral electrochemical luminescence sensors [124]. In 2018, Wu et al. synthesized homoleptic cyclometalated iridium(III) complex nanowires and used them as luminescent receptors modified into an indium tin oxide (ITO) sensor and added the coreactant tripropylamine to the system to optimize the luminescence system [125]. Due to the differences in electronic transmission capacity between different free radicals, ECL sensors are used for the high-performance recognition of proline enantiomers. As the cyclometalated iridium(III) complex displays abundant three-wire states and excellent electrochemical properties, it has great application potential in ECL sensors. In 2019, Zhu et al. used cadmium-carbonate-modified $g\text{-C}_3\text{N}_4$ to construct a nanocomposite material as the luminescent receptor and then produced different degrees of ECL signal quenching as a result of the host-guest interaction with propranolol enantiomers to achieve specific recognition [126]. Among the various carbon nanomaterials, $g\text{-C}_3\text{N}_4$, as a typical polymer semiconductor, can effectively activate molecular oxygen to generate superoxide radicals, which is conducive to the construction of ECL sensors. In 2021, Zhao et al. functionalized Ag_2S quantum dots with N-acetyl-L-cysteine to prepare chiral nanocomposites and solidified them on the surface of two-dimensional C_3N_4 -modified electrodes to serve as chiral recognition units and luminescent receptors [127]. The selective recognition of tyrosine enantiomers can be achieved through supramolecular interactions. The special combination of chiral molecules and quantum dots creates more possibilities for the construction of chiral electrochemical luminescence materials.

ECL sensors consist of a three-electrode system similar to that of electrochemical sensors. The macrocyclic compounds solidified on the electrode surface are also conducive to

supramolecular molecular recognition. In 2021, Liu et al. synthesized a flexible molecular universal joint (DPMUJ) based on functionalized chiral pillar[5]arenes with bipyridine and then coordinated the DPMUJ enantiomers with luminous ruthenium ions to form chiral receptors with an ECL signal response [128]. Experiments have shown that the chiral host can selectively recognize ECL signals of different intensities after interactions with amino acid enantiomers. A novel method to synthesize supramolecular assemblers with chiral recognition ability was reported, providing new guidelines for designing chiral ECL sensors constructed with macrocyclic compounds. In 2022, Wu et al. activated an achirality macrocyclic compound, benzo[3]uril, to produce a chiral microenvironment through supramolecular interactions with chiral guest molecules. Since benzo[3]uril produces an ECL signal response, the ECL signal changes can be directly generated through the competitive interaction between chiral guest molecules and macrocyclic lumen [129]. Eventually, the specific recognition of L-penicillamine enantiomers was achieved (Figure 16). Therefore, a simple and universal method is used to construct chiral sensors and a series of ECL sensors for molecular recognition.

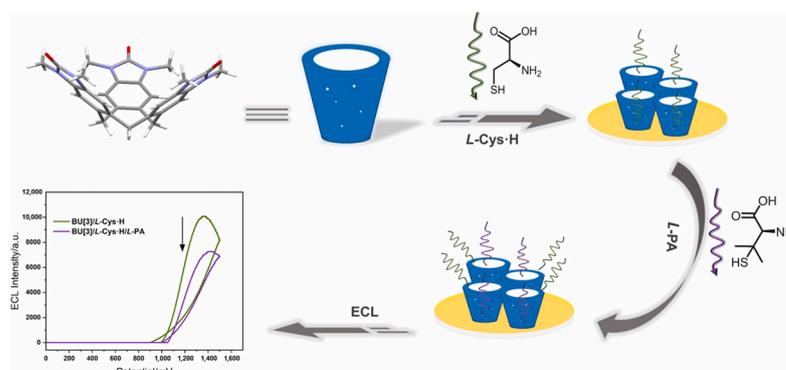


Figure 16. Schematic diagram of ECL chiral sensor construction and recognition of thyroxine enantiomer. Adapted from Ref. [129]. Copyright 2022 ELSEVIER.

5. Photoelectric Chemical Chiral Supramolecular Sensors

Photoelectric chemical sensors represent a new technology in which light is used as the excitation source, and photocurrent or photovoltage is used as the detection signal to quantitatively analyze the relationship between the target and the photocurrent or photovoltage for electrochemical and biometric identifications [130]. The photochemical process refers to the transfer of charge generated through electron excitation due to photon absorption by molecules, ions, and semiconductor materials, thus achieving the conversion of light energy into electric energy [131]. According to the operating principle of the sensor, light is irradiated on the photoactive material, the electrons in the photoactive material become excited under irradiation, and the recognition probe on the material captures the target analyte, leading to the change in photocurrent or photovoltage [132]. Although the application of PEC technology in the field of molecular recognition has become a research hotspot, it is still hard to find a suitable chiral photoreceptor.

Since the constructed PEC sensor used in analysis and detection is still a common three-electrode system, in this review, we focus on the design of photosensitive electrode materials. In 2020, Liu et al. synthesized a titanium dioxide nanochannel array as a photosensitive substrate and then modified β -cyclodextrin as a chiral recognition unit by coupling it on the nanomaterial and ultimately achieved photoelectric chemical detection triggered through ion current rectification with the composite nanomaterial-modified working electrode [133]. The constructed sensor enabled the specific recognition of L-histidine enantiomers. In 2021, Zhou et al. reported the construction of an L-histidine-functionalized pure chiral metal-organic skeleton using titanium dioxide nanotubes [134]. Then, by studying host-guest interactions, they found that the photocurrent signal response intensity of the nanocomposite to L-dopa was significantly lower than that of D-dopa, thus achieving

the selective recognition of dopa enantiomers. In 2021, Yang et al. reported the preparation of chiral nanofibers through the electropolymerization of thiophene-modified pillar[5]arene monomers [135]. The obtained polymer showed a significant circular dichroism signal, and the constructed photoelectric chemical sensor could selectively identify ascorbic acid enantiomers. In 2021, Zhao et al. used graphitic carbon nitride quantum dots as the photocurrent material and solidified them on the ITO electrode, and then modified the layer-by-layer of chiral multifarene[3,2,1] as the chiral recognition unit [136]. Through supramolecular interaction between chiral macrocyclic cavities and guest molecules, the photocurrent decreases to varying degrees. A super-sensitive chiral photoelectric sensor was constructed to selectively recognize thyroxine enantiomers (Figure 17).

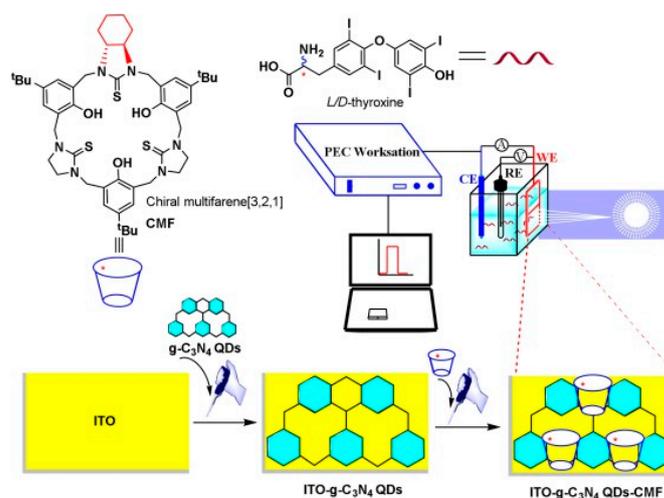


Figure 17. Schematic diagram of PEC chiral sensor construction and recognition of thyroxine enantiomer. Adapted from Ref. [136]. Copyright 2021 ELSEVIER.

6. Conclusions

The analysis results of various methods used for detecting materials in the above-mentioned reports are summarized in Table 1. The detection range and detection limit are also listed in Table 1. We intuitively compared the characteristics of different analytical methods and the advantages of different strategies for the construction of sensors. The electrochemical analysis methods mostly have wider detection ranges and lower detection limits than optical analysis methods (Table 1). However, few reports exist on the construction of ECL and PEC sensors, the general detection range is over several orders of magnitude, and the detection limit can reach 1.0 nM or lower. Therefore, ECL and PEC detection methods have certain application potential.

In early chiral supramolecular systems, many studies focused on the advantages of supramolecular methods and the structural diversity of components but ignored the relevant studies on the mechanism and properties of chiral transfer, and as a result, many control factors of microchirality have not been developed [137]. In the last five years, the research on chiral supramolecular sensors has made a qualitative leap. Researchers have examined more methods of supramolecular chiral memory and regulation, opened up more new perspectives on molecular recognition mechanisms, and prompted research enthusiasm and extensive application of supramolecular chiral effect in this field. There are many chiral supramolecular sensors for detecting small amino acid enantiomers, but there are few sensors for the specific recognition of chiral drugs, indicating that chiral supramolecular sensors cannot analyze complex chiral molecules, and a huge gap exists in the fabrication process of these sensors. Although the relevant research on chiral supramolecular sensors has been productive, the functionalization of materials is still challenging, and more methods are needed to achieve the biological compatibility of materials. Finally, chiral supramolecular sensors have broad application prospects for the detection of chiral drugs

in organisms and chiral pollutants in the environment, as well as great potential for the development of intelligent chiral sensing systems and their commercial application.

Table 1. Comparison of chiral supramolecular sensor analysis for identification of enantiomers.

Detection Method	Targeted Substrates	Linea Range (μM)	Lod (nM)	References
Fluorescence	D-penicillamine	$30.0\text{--}2.0 \times 10^3$	5.0×10^3	[61]
Fluorescence	D-penicillamine	23.0–0	80.0	[62]
Fluorescence	D-penicillamine	$2.5 \times 10^{-2}\text{--}0.7$	8	[63]
Fluorescence	L/D-arginine	0–0.4/0–0.3	1.9/1.8	[64]
Fluorescence	D-aspartic	3.0–31.0	14.3	[65]
Fluorescence	L-morphine	$2.8 \times 10^{-2}\text{--}0$	60	[67]
Fluorescence	L/D-phenylalaninol	$1.0 \times 10^2\text{--}1.0 \times 10^4$	$3.02 \times 10^2/3.97 \times 10^3$	[68]
Fluorescence	L-histidine	$1.0 \times 10^{-7}\text{--}1.8 \times 10^{-6}$	8.0×10^{-5}	[70]
CD	L/D-histidine	5.0–50.0	1.0×10^4	[75]
Colorimetric	D-cysteine	$5.0\text{--}1.0 \times 10^2$	4.9×10^3	[79]
RTP (solution/nanopaper)	L-lysine	$0.0\text{--}2 \times 10^4/0.0\text{--}2.0 \times 10^3$	0.30/0.97	[80]
SPR	L-tryptophan	$0.15 \times 10^3\text{--}2.5 \times 10^3$	0.1×10^6	[81]
CV	D-cysteine	$1.0 \times 10^{-2}\text{--}1.8$	8.7	[90]
DPV	L-cysteine	$1.0 \times 10^{-7}\text{--}1.0 \times 10^{-1}$	6.0×10^{-6}	[95]
DPV	D-carnitine	$3.6 \times 10^{-10}\text{--}4.0 \times 10^{-7}$	2.2×10^{-7}	[96]
DPV	esomeprazole (ESOM)	$1.0 \times 10^{-8}\text{--}2.0 \times 10^{-7}$	1.9×10^{-6}	[97]
DPV	(1s,2s)- rseudoephedrine	$1.0 \times 10^{-9}\text{--}1.0 \times 10^{-8}$	2.9×10^{-7}	[98]
DPV	L/D-tryptophan	$0.5\text{--}1.7 \times 10^2$	9.6/38.0	[102]
DPV	L/D-phenylalanine	$10.0\text{--}5.0 \times 10^3$	27.0/52.0	[103]
DPV	L/D-tryptophan	$10.0\text{--}5.0 \times 10^3$	63.0/3.5	[104]
DPV	L-phenylalanine	0.2–13.0	80.0	[105]
DPV	L/D-tyrosine	$10.0\text{--}1.0 \times 10^3$	$4.81 \times 10^3/6.89 \times 10^3$	[106]
DPV	L/D-tryptophan	$1.0 \times 10^2\text{--}4.0 \times 10^3$	$1.85 \times 10^4/1.34 \times 10^4$	[107]
DPV	R/S-NaX	0.4–6.0	70.0	[108]
DPV	L/D-tryptophan	$10.0\text{--}0.5 \times 10^3$	9.8/23.0	[109]
DPV	L/D-penicillamine	$10.0\text{--}0.5 \times 10^3$	18.0/79.0	[109]
DPV	L/D-tryptophan	$40.0\text{--}4.0 \times 10^3$	$2.8 \times 10^3/3.7 \times 10^3$	[112]
DPV	L/D-tryptophan	$10.0\text{--}5.0 \times 10^3$	24.0/55.0	[117]
DPV	L-tyrosine	$10.0\text{--}1.6 \times 10^3$	-	[119]
ECL	L/D-proline	$10.0\text{--}1.0 \times 10^5$	1.0	[125]
ECL	R/S-propranolol	$1.0\text{--}1.0 \times 10^3$	3.3×10^2	[126]
ECL	L-penicillamine	$1.0 \times 10^{-4}\text{--}10.0$	1.0×10^{-3}	[129]
PEC	L-histidine	$0.2 \times 10^3\text{--}1.0 \times 10^3$	6.75×10^4	[133]
PEC	L/D-DOPA	$1.0\text{--}10.0/20.0\text{--}1.0 \times 10^2$	2.4×10^2	[134]
PEC	L-ascorbic acid	0.1–0	2.44	[135]
PEC	L/D-thyroxine	$1.0 \times 10^{-4}\text{--}1.0 \times 10^{-2}$	$6.7 \times 10^{-2}/8.5 \times 10^{-2}$	[136]

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